

Oscillatory Exchange Coupling and Positive Magnetoresistance in Epitaxial Oxide Heterostructures

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Oscillations in the exchange coupling between ferromagnetic $La_{2/3}Ba_{1/3}MnO_3$ layers with paramagnetic $LaNiO_3$ spacer layer thickness has been observed in epitaxial heterostructures of the two oxides. This behavior is explained within the RKKY model employing an *ab initio* calculated band structure of $LaNiO_3$, taking into account strong electron scattering in the spacer. Antiferromagnetically coupled superlattices exhibit a positive current-in-plane magnetoresistance.

75.70.Cn, 75.30.Vn, 71.18.+y, 75.70.Pa

Since the discovery of giant magnetoresistance [1] and oscillatory interlayer coupling [2], metallic magnetic multilayers have been the subject of intensive research [3]. A physical picture of the coupling is provided in terms of quantum interference due to confinement of electrons in the nonmagnetic spacers [4,5]. Ruderman-Kittel-Kasuya-Yosida (RKKY) theory [6], successfully used to describe the effect, appears to be a limiting case of a more general approach [5]. The prediction that oscillations periods are determined by the extremal spanning vectors of the Fermi surface of the nonmagnetic spacer is now supported by a wealth of experimental data [7]. The phase and magnitude of the coupling oscillations apparently are sensitive to the interface matching of the electron bands for a particular magnetic configuration of the layers [3–5].

While these effects have been studied in many simple metal and alloy systems, little progress has been achieved in investigating them in multilayers consisting entirely of compounds [8]. In this Letter we report the first observation of oscillatory coupling in heterostructures fabricated entirely of oxides, thus extending the field to a novel class of materials. The ferromagnetic layer material, barium-doped lanthanum manganese oxide $La_{2/3}Ba_{1/3}MnO_3$ belongs to the family of metallic manganese oxides that exhibit very large (colossal) magnetoresistance [9]. Double exchange ferromagnetism [10] predicts half-metallicity in these compounds, which has been justified by *ab initio* band structure calculations [11] as well as confirmed (to a certain degree) experimentally [12]. As a spacer layer material, we have employed $LaNiO_3$, which is lattice-matched to $La_{2/3}Ba_{1/3}MnO_3$, and is the only rare earth nickelate that is a paramagnetic metal [13]. Its susceptibility is strongly enhanced by electron-electron exchange interactions [14]. In thin film form, it is a better conductor (resistivity of $50 - 100 \mu\Omega \cdot cm$) than the manganites (resistivity of $300 - 500 \mu\Omega \cdot cm$). Although advances in oxide film growth permit the fabrication of atomically defined lay-

ered structures with nanometer scale periodicity [15], the very strict control over stoichiometry and deposition conditions required to produce multilayers remains a significant experimental challenge.

In our previous paper [16] we presented the evidence for antiferromagnetic (AFM) interlayer coupling in this system. Here we demonstrate that the initially AFM coupling becomes ferromagnetic (FM) at larger spacer thicknesses and explain this behavior within the RKKY model.

Superlattices were grown on heated $SrTiO_3(001)$ substrates by block-by-block molecular beam epitaxy (MBE) [17] in the presence of an ozone flux, using previously described procedure [18]. The RHEED patterns observed during the block-by-block deposition, indicated smooth surfaces at the completion of each monolayer throughout the entire process of growth. Characterization by high resolution X-ray diffraction showed that the films were monocrystalline with *c*-axis oriented perpendicular to the substrate. Well-defined satellite peaks indicated the sharpness of the interfaces and good superlattice periodicity [16]. Cross-sectional transmission electron microscopy demonstrated the continuity of the layers over macroscopic scales, and notably defect-free structures on atomic length scales.

For a single very thin manganite film encapsulated between two nickelate layers, hysteresis loops with the field applied in various directions in the (001) plane of the film can be described by a biaxial magnetocrystalline anisotropy. This demonstrates that domain-wall pinning is negligible and justifies a model of coherent rotation of the magnetization in the analysis of hysteresis.

Figure 1 shows typical M-H curves for a series of $(La_{2/3}Ba_{1/3}MnO_3 / LaNiO_3)_{10}$ (001) superlattices. The thickness of each manganite layer was fixed at 12 unit cells (u.c.), while that of nickelate varied from 3 u.c. to 10 u.c. [19]. For protection, the structures were encapsulated between two 50Å-thick nickelate layers.

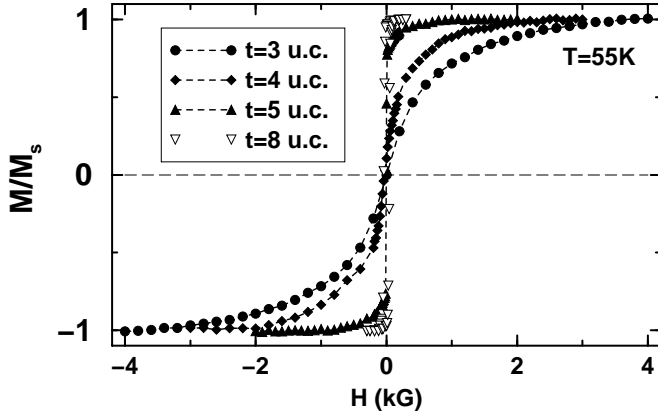


FIG. 1. Hysteresis loops for a series of $(La_{2/3}Ba_{1/3}MnO_3$ 12 u.c. / $LaNiO_3$ t)₁₀ superlattices obtained using a superconducting susceptometer.

The magnetic field was directed along the [100] direction, coinciding with the easy axis of the manganite films. Samples with spacer layers three- and four-unit-cell thick, showed zero remanence and high saturation fields typical of AFM coupling. The spontaneous magnetizations of these samples at low temperatures were less than one percent of saturation, illustrating that all the periods have the same thickness and composition, and that there were no pinholes bridging between layers. However, the shape of the magnetization curves suggests a substantial biquadratic contribution to the interlayer coupling [20]. For thicker spacer structures, hysteresis loops had high remanence and the saturation fields were much lower. The coupling constants were estimated by fitting hysteresis curves with those calculated from a model with coherent rotations [21].

To estimate the coupling constants of small magnitude anticipated for structures with thicker spacer layers, a series of samples with varied spacer layer thickness t were grown in a "spin-valve" [22] four-layer geometry: $La_{2/3}Ba_{1/3}MnO_3$ 12 u.c. / $LaNiO_3$ t / $La_{2/3}Ba_{1/3}MnO_3$ 12 u.c. / $La_{1/3}Ca_{2/3}MnO_3$. In this configuration the magnetization of one manganite layer is free to rotate while the other is pinned by the 150Å-thick epitaxial layer of the antiferromagnet, $La_{1/3}Ca_{2/3}MnO_3$ [23]. Measurements were performed on field-cooled films with a magnetic field applied along the [100] direction. As an example, a hysteresis loop of a structure with an eight-unit-cell thick spacer is shown in Fig. 2. The inset shows the hysteresis loop corresponding to an unbiased FM layer ("minor" loop). The magnetic coupling between the manganite layers leads to a change of an external field at which the magnetization of the unbiased layer flips. This results in a shift of the minor loop along the field axis, the sign and magnitude of the shift being a quantitative measure of the interlayer coupling. No such displacement was detected for samples with thicker spacers, demonstrating that the magnetic layers are decoupled. For nickelate spacers six- and seven-

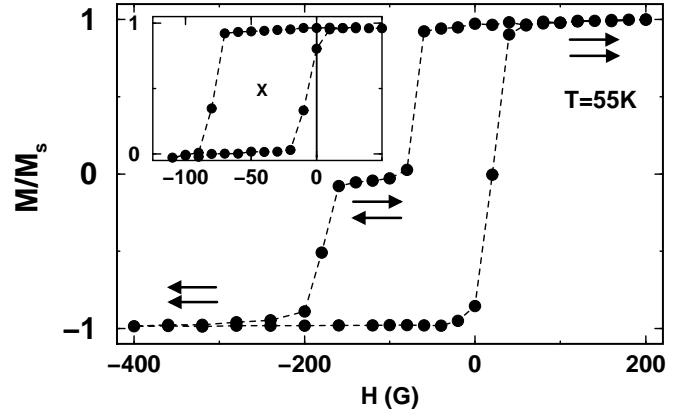


FIG. 2. Hysteresis loop for a "spin-valve"-like structure with eight-unit-cell thick $LaNiO_3$ spacer obtained using a superconducting susceptometer. Arrows indicate magnetization directions. Inset: minor hysteresis loop.

unit-cell thick single hysteresis loops were observed with the magnetizations of both layers reversing their orientations simultaneously. This is indicative of a strong FM interaction. A lower limit on the strength of the coupling can be set by the difference of the coercive fields of the biased and free FM layers. In agreement with the superlattice data, the hysteresis loop of the structure with a thin (four-unit-cell thick) spacer layer reflected the presence of strong bilinear AFM and biquadratic couplings.

The combined data from both sets of measurements were used to obtain the variation of bilinear coupling strength J_1 with spacer layer thickness (curve a in Fig. 3) [24]. As one can see, the initially AFM coupling changes sign before vanishing. The coupling strength is comparable to that found in many conventional systems.

A conventional RKKY model with additional damping caused by strong electron scattering in the nonmagnetic

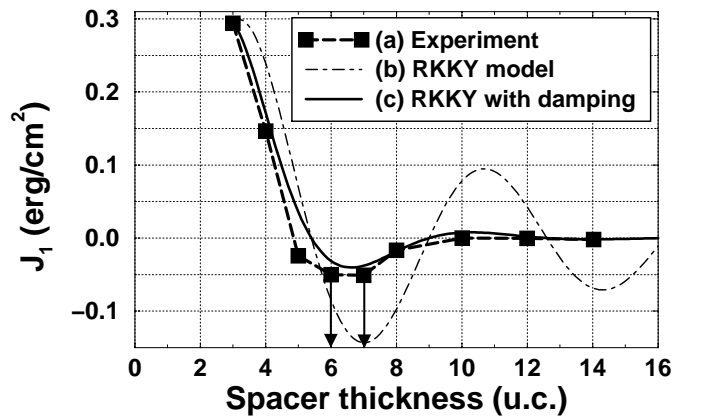


FIG. 3. Spacer thickness variation of experimental (curve a) and calculated (curves b and c) coupling strength. The theoretical curve is $J_1(t) = A \cos(2\pi t/T_{LP})/t \cdot \exp(-t/\lambda)$ with long period $T_{LP} = 7.2$ u.c. and damping length $\lambda = 3$ u.c. as well as without damping ($\lambda = \infty$). The amplitude A is chosen to match the experimental value at $t = 3$ u.c.

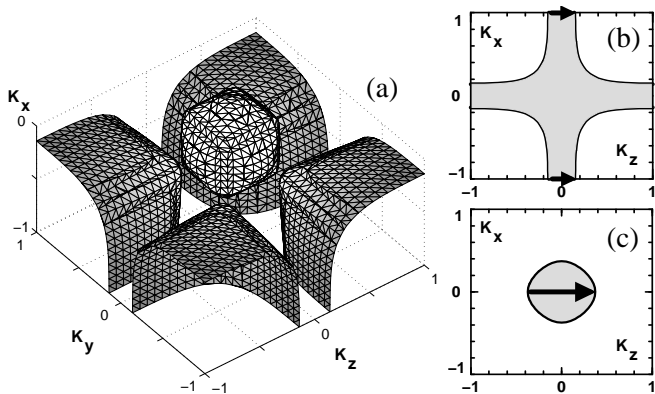


FIG. 4. (a) Fermi surface of $LaNiO_3$ consists of a small Γ -point electron sheet and a huge R -point hole pocket (half of it is displayed). (b) $k_y = \pi/a_0$, (c) $k_y = 0$ critical cross-sections of the Fermi surface. Arrows indicate extremal spanning wave vectors. The wave vectors are in units of π/a_0 .

layer has been used to describe the observed variation of the coupling. The topography of the Fermi surface of the nickelate spacer is vital to the description of the exchange coupling. The band structure and density of states of $LaNiO_3$ have been computed using the pseudo-potential plane wave approach [25], and are in good agreement with previous calculations [29]. The Fermi surface of $LaNiO_3$ is depicted in Fig. 4a. For the [001] direction, there are two apparent extremal spanning vectors associated with long $T_{LP} = 7.2$ u.c. (Fig. 4b) and short $T_{SP} = 2.7$ u.c. (Fig. 4c) oscillation periods. Due to a much larger radius of curvature of the Fermi surface K_F at the long period extremal spanning vector, its contribution is dominant. In addition, short periods are expected to be suppressed by interface roughness. Because of the nested-like feature of the Fermi surface, the asymptotic RKKY formula is not applicable for thin ($t < K_F a_0^2 \sim 40$ u.c.) spacers. Considerations similar to ones used in the case of complete planar nesting [6] significantly modify the spacer thickness dependence of the coupling strength which is in this case given by: $J(t) \propto -\cos(2\pi t/T_{LP})/t$, where t is the spacer thickness. The comparison with experimental data (curves a and b in Fig. 3) reveals good agreement in both period and phase. However, the experimentally observed decay rate of the coupling strength is faster than predicted. We argue that strong electron scattering in $LaNiO_3$ may account for the oscillation damping of the form of $\exp(-t/\lambda)$, where λ is the scattering length of the "coupling" electrons. An estimate for $\lambda = 3$ u.c. [30] obtained from the measured resistivity describes the observed damping reasonably well (see curve c in Fig. 3) [31].

Current-in-plane magnetoresistance (MR) measurements were made using a standard four-probe DC technique. Results are shown in Fig. 5 for three superlattices with spacer layer thicknesses of three, four, and six unit cells, respectively. The observed decrease in resistance at large fields originates from the negative MR of the

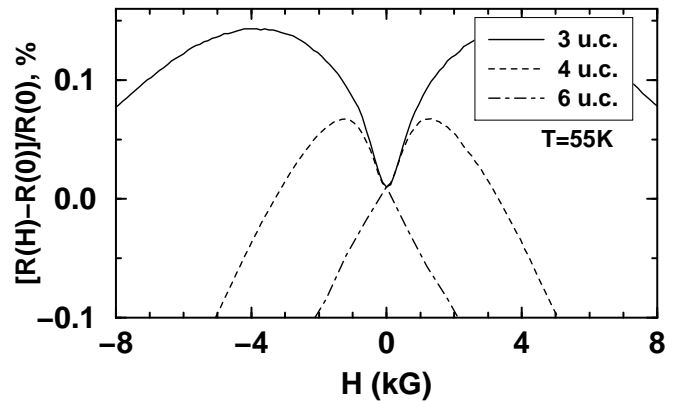


FIG. 5. Magnetoresistance vs in-plane magnetic field dependence for the $(La_{2/3}Ba_{1/3}MnO_3 \text{ 12 u.c.} / LaNiO_3 \text{ } t)_{10}$ superlattices. Actual magnetoresistance is higher since part of the current is shunted by $LaNiO_3$ capping layers.

manganite layers. Unlike the case of bulk material and thicker films, substantial negative MR is observed in single ultrathin manganite films well below the Curie temperature [35]. The low-field positive MR is present only in structures that exhibit AFM interlayer coupling. The resistance has a minimum for the AFM configuration, whereas a maximum is seen when the layers become FM aligned. This provides compelling evidence that the phenomenon is associated with the configuration of the magnetizations of adjacent layers. Measurements performed with a field applied at various angles to the direction of current yielded the same result, thus demonstrating the effect is not caused by in-plane MR anisotropy. The positive MR we found is in contrast with the negative magnetoresistance conventionally observed in transition metal multilayers. In latter structures the effect of the positive sign is only observed in systems with spin asymmetries in the electron scattering in successive FM layers [36].

We speculate that this behavior may be caused by the intrinsic negative MR of the manganite layers. In this model, an indirect exchange interaction affects the transport in the manganites [37]. If one views the superlattice as a parallel resistor network, the low-field positive MR can indeed be achieved if the coupling is AFM with a biquadratic contribution. Alternatively, the effect may originate from the variation of the electronic structure of the system due to the change in the magnetic configuration.

In conclusion, heterostructures consisting of layers of a colossal magnetoresistance material separated by a non-magnetic metallic oxide spacers have been successfully grown by MBE method. The high quality of the heterostructures allowed us for the first time to study the magnetic interlayer coupling, and demonstrate the oscillatory dependence of the coupling strength on the spacer thickness. Exchange coupling in this exotic system has been described within the RKKY model. The origin of the positive magnetoresistance with a cusp-like feature at small magnetic fields remains unclear at this time.

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